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Linear and nonlinear absorption coefficients of spherical two-electron quantum dot

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ABSTRACT

In this study, optical properties of two-electron quantum dot confined by an infinite spherical potential surface have been investigated. Linear, nonlinear and total absorption coefficients of S → P, P → D and D → F dipole-allowed transitions between singlet–singlet and triplet–triplet states have been calculated as a function of dot radius and photon energy. The results show that the change of dot radius and incident optical intensity effects the peak positions and amplitudes of linear and nonlinear absorption coefficients. Besides, it has been found that the absorption coefficients of transitions between triplet states are stronger than those of the singlet states, and also triplet absorption transitions occur at higher energies.

1. Introduction

In the past decades, various theoretical studies on quantum dots, where electrons and holes are confined in all three dimensions, have attracted an increasing interest. The confinement of electron in Quantum Dots (QDs) leads to significant changes in optical properties and many novel features, not found in the bulk materials. QDs are known as artificial atoms because of having shell structures and discrete energy levels, and QDs play an important role in optoelectronic and microelectronic devices. Therefore, many authors have studied the binding energies, the effects of electric and magnetic field and other physical properties of one-electron QD with various size and shapes [1–6]. In one-electron QDs, linear and nonlinear absorption coefficients have attracted the attention of researchers both experimentally and theoretically [7–25], because QDs have the potential for the device applications such as far-infrared photodetectors, laser amplifiers (wavelength ∼ 10 μm), optical memories, light emitting diodes and high-speed electro optical modulators [26–28].

Recently there is an increasing interest to QDs with two-electrons (the confined helium and helium-like ions, that is Li^{+}, Be^{++}, C^{+++} etc.). The Coulomb interaction between electrons effects considerably the physical properties of such structures and must be considered in calculations [29]. Therefore, various properties of the confined helium and helium-like ions have been investigated by many researchers [30–38]. In the work of Ndengué and Motapon [31] were computed the energy levels of the ground and a few excited states of helium and helium-like ions confined in a finite potential well by using configuration interaction method. Flores-Riveros and Rodríguez-Contreras [33] and Yakar et al. [35] carried out the ground and excited energy states for the confined helium and helium-like ions spherically enclosed by an impenetrable box. They also calculated the ionization energies and the ionization dot radii of the confined systems. As is well known, ionization energy is defined as that necessary to remove an electron from the outermost orbital of an atom (or the confined atom, QD) [33]. As for optical properties of two-electron QDs (2e-QDs), the absorption coefficients were calculated by Huang and Libin [39] by using the exact diagonalization techniques and the compact density-matrix approach. Şahin [40] studied a detailed investigation of the linear and nonlinear optical properties by utilizing the full numeric matrix diagonalization technique. Similarly, Lu et al. [41] investigated the optical absorption coefficients and refractive index changes in a GaAs/AlGaAs QD by using compact-density matrix approach and exact analytical method. Very recently, the optical properties of 2e-QDs with a Gaussian confinement potential were analyzed by Coden et al. [42]. Above mentioned theoretical studies related to the QD with two-electron are focused on the calculation of optical transition

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between the lowest energy state, which is S → P singlet transition of the ground ($L = 0$) and the first excited ($L = 1$) states. Therefore, the theoretical studies on the absorption coefficients of 2e-QDs are still important, especially in transitions among higher excited electronic states. To our knowledge, up to now the linear and nonlinear absorption coefficients of optical transitions among the excited energy states in 2e-QD have not been studied in literature.

In this paper, we have calculated the wavefunctions and the energy eigenvalues of $1s^2$ (S), 1s2p (P), 1s3d (D) and 1s4f (F) singlet and triplet states of spherical 2e-QD. In addition, we have also investigated the linear, third-order nonlinear and total absorption coefficients of the S → P, P → D and D → F optical transitions between singlet–singlet and triplet–triplet states as a function of dot radius and photon energy. We used the GaAs material in our calculations.

2. Theory and definition

In the effective mass approximation, the electronic Hamiltonian of a 2e-QD confined by an infinite spherical symmetric potential well can be written, in atomic units (au), as follows:

$$H = \sum_{i=1}^{2}\left(-\frac{\nabla_i^2}{2m^*} - \frac{Z}{\varepsilon r_i} + \sum_{i<j}^{2}\frac{1}{\varepsilon r_{ij}}\right) + V_c\,(r_1, r_2)\,, \tag{1}$$

in which Z is the impurity charge, r_i is the distance between ith electron and impurity, r_{ij} is the distance between the electrons, m^* and ε are the relative effective mass of electron and the relative dielectric constant of medium. The first two terms of the Hamiltonian are one-electron operators and the third term is two-electron operator. The last term $V_c\,(r_1, r_2)$ is the confinement potential and it is defined as

$$V_c(r_1, r_2) = \begin{cases} 0, & \text{if } r_1 \text{ and } r_2 < R \\ \infty, & \text{if } r_1 \text{ or } r_2 \geqslant R \end{cases}, \tag{2}$$

in which R is dot radius. The Schrödinger equation of a two-electron system is given by

$$H\psi(q_1, q_2) = E\psi(q_1, q_2), \tag{3}$$

in which E and ψ are the energy eigenvalue and the eigenfunction of Hamiltonian operator, and q_i is the coordinate of ith electron. The wavefunctions $\psi(q_1, q_2)$ being an eigenfunction of $\mathbf{S^2}$ and $\mathbf{S_z}$, which are the square of total spin angular momentum operator $\mathbf{S}$ and its z-component, satisfy the normalization. In a two-electron system, the wavefunction ψ can be expressed as a Slater determinant or a linear combination of two Slater determinants, which are consistent of one electron spin orbitals $u_\mu(q_i)$ containing the spatial $\phi_{nlm}(\mathbf{r})$ and spin part $\sigma_{\mathbf{m_s}}(s)$, μ denotes quantum numbers $nlmm_s$. The wavefunction must provide the boundary conditions, that is, $\psi(r_1, R) = \psi(R, r_2) = \psi(R, R) = 0$, and at the same time the spatial part of one-electron spin orbital $\phi_{nlm}(\mathbf{r})$ must be also satisfied the same boundary conditions at $r = R$.

The electronic states of n-electron system are shown as ^{2S+1}L, where S and L are the total spin and orbital quantum number of system. In a two-electron system, the total spin S is either 0 or 1, corresponding to the singlet and the triplet states. The wavefunction expressed as product of spatial and spin part must be antisymmetric. Therefore, in singlet case, the spatial part of wavefunction is symmetric since the spin part is antisymmetric. This case is reverse for the triplet case.

For a two-electron system, the Slater determinant wavefunction of the $1s^2$ ground state, $\psi\,(q_1, q_2)$, can be expanded over one-electron orbitals as follows:

$$\psi\,(q_1, q_2) = 2^{-1/2}\,(\phi_{1s}(\mathbf{r}_1)\phi_{1s}(\mathbf{r}_2))\,[\alpha(1)\beta(2) - \beta(1)\alpha(2)]\,, \tag{4}$$

where $\alpha(i)$ and $\beta(j)$ denote the spin-up and spin-down functions, respectively. For the 1s nl excited configuration of a two-electron system, $m = 0$, the wavefunctions which are expansion of Slater determinants can be written as [43]:
for singlet state

$$\begin{aligned}\psi\,(q_1, q_2) \;=\; & 2^{-1/2}\,[\phi_{1s}(\mathbf{r}_1)\phi_{nl}(\mathbf{r}_2) + \phi_{nl}(\mathbf{r}_1)\phi_{1s}(\mathbf{r}_2)] \\ & \times 2^{-1/2}\,(\alpha(1)\beta(2) - \beta(1)\alpha(2))\,,\end{aligned} \tag{5}$$

and for triplet states

$$\begin{aligned}\psi(q_1, q_2) &= 2^{-1/2}\,[\phi_{1s}(\mathbf{r}_1)\phi_{nl}(\mathbf{r}_2) - \phi_{nl}(\mathbf{r}_1)\phi_{1s}(\mathbf{r}_2)]\;\; \alpha(1)\alpha(2) \\ \psi(q_1, q_2) &= 2^{-1/2}\,[\phi_{1s}(\mathbf{r}_1)\phi_{nl}(\mathbf{r}_2) - \phi_{nl}(\mathbf{r}_1)\phi_{1s}(\mathbf{r}_2)]\;\; \beta(1)\beta(2) \\ \psi(q_1, q_2) \;=\; & 2^{-1/2}\,[\phi_{1s}(\mathbf{r}_1)\phi_{nl}(\mathbf{r}_2) - \phi_{nl}(\mathbf{r}_1)\phi_{1s}(\mathbf{r}_2)] \\ & \times 2^{-1/2}\,(\alpha(1)\beta(2) + \beta(1)\alpha(2))\,.\end{aligned} \tag{6}$$

If one-electron orbitals are normalized, $m = 0$, the singlet energy E^{S}, the triplet energy E^{T} and the average energy E^{av}, which is equal to sum of the state energies divided by number of states of the $1sn\ell$ configuration, are given by

$$E^{\mathrm{S}} = I(1s) + I(n\ell) + J(1s, n\ell) + K(1s, n\ell) \tag{7}$$

$$E^{\mathrm{T}} = I(1s) + I(n\ell) + J(1s, n\ell) - K(1s, n\ell) \tag{8}$$

$$E^{\mathrm{av}} = I(1s) + I(n\ell) + J(1s, n\ell) - \frac{1}{2}K(1s, n\ell), \tag{9}$$

in which $n\ell$ are the arbitrary quantum numbers, $I(n\ell)$ is one electron integral, $J(1s, n\ell)$ and $K(1s, n\ell)$ are the electron–electron Coulomb and Exchange energy integrals respectively. For the ground state ($1s^2$), these integrals are modified as $n\ell = 1s$ and $K(1s, 1s) = 0$. These energy integrals in au are given as follows:

$$I(n\ell) = \left\langle \phi_{n\ell}(\mathbf{r}) \left| -\frac{\nabla^2}{2m^*} - \frac{Z}{\varepsilon r} \right| \phi_{n\ell}(\mathbf{r}) \right\rangle, \tag{10}$$

$$J(1s, n\ell) = \left\langle \phi_{1s}(\mathbf{r}_1)\phi_{n\ell}(\mathbf{r}_2) \left| \frac{1}{\varepsilon r_{12}} \right| \phi_{1s}(\mathbf{r}_1)\phi_{n\ell}(\mathbf{r}_2) \right\rangle, \tag{11}$$

and

$$J(1s, n\ell) = \left\langle \phi_{1s}(\mathbf{r}_1)\phi_{n\ell}(\mathbf{r}_2) \left| \frac{1}{\varepsilon r_{12}} \right| \phi_{n\ell}(\mathbf{r}_1)\phi_{1s}(\mathbf{r}_2) \right\rangle. \tag{12}$$

In order to compute the energies given in Eqs. (7)–(9), one should calculate one- and two-electron molecular integrals appearing in HFR approximation. For QDs, one- and two-electron integrals given in Eqs. (10)–(12) can be easily evaluated by modifying for appropriate consideration of the boundaries in the expressions of atomic systems [44].

The $\phi_k(\mathbf{r})$ may be written as a linear combinations of unnormalized Slater Type Orbitals (STOs), χ, as follows:

$$\phi_k(\mathbf{r}) = \sum_{\sigma=1}^{t} c_{\sigma k}\chi_k(\mathbf{r}, \zeta_\sigma), \tag{13}$$

where t is the number of basis sets.

Absorption Coefficients (ACs) are one of the most important parameters of the optical transitions. As the electromagnetic radiation passes through QD, the ACs give a measure of the decrease ratio in the radiation intensity. The ACs $\alpha(\omega)$ can be evaluated from the susceptibility $\chi(\omega)$ term. For a spherical QD with two-electron, the linear $\alpha^{(1)}(\omega)$ and the third-order nonlinear $\alpha^{(3)}(\omega, I)$ ACs are defined by [39,40,45]

$$\alpha^{(1)}(\omega) = \omega\sqrt{\frac{\mu}{\varepsilon_r}}\;\frac{\rho\,\hbar\Gamma_{fi}}{\left(E_{fi} - \hbar\omega\right)^2 + \left(\hbar\Gamma_{fi}\right)^2}\;\left\langle M_{fi}\right\rangle^2, \tag{14}$$